Effect of the driving frequency on an atmospheric pressure RF capacitively coupled plasma in Argon

M Atanasova(*)[1,2], E A D Carbone[3], G Degrez[1] and J J A M van der Mullen[3]

[1]*Department of Applied Sciences, ULB, 50 av. Fr. Roosevelt – CPI 165/04, Brussels, Belgium*
[2]*Department for Language Teaching and International Students, Sofia University, 27 Kosta Loulchev St., 1111 Sofia, Bulgaria*
[3]*Department of Applied Physics, Eindhoven University of Technology, P.O. Box 513, 5600 MB Eindhoven, The Netherlands*
(*) matanaso@ulb.ac.be

The plasma properties of a capacitively coupled atmospheric pressure plasma in Argon as a function of the driving frequency f_d is studied for a number of frequencies in the range 4–40 MHz by means of a 2D fluid model. It is observed that the increase of f_d while keeping the applied voltage constant results into amplification of the chemical processes. More neutral active species are produced, which is mainly due to the increase of the electron density n_e. However the increase of n_e will also facilitate the destruction of excited species Ar^* so that the increase of excited species density $n(Ar^*)$ will lag behind that of n_e.

Capacitively coupled atmospheric pressure plasmas are widely used to produce cold atmospheric plasmas (CAPs) while still delivering large fluxes of active species useful for applications. They offer a variety of possibilities for treatment of sensitive surfaces including biological ones as well as many other applications. To characterize their operation and establish the plasma properties as a function of the driving frequency f_d, a CAP in argon is studied theoretically by means of a time-dependent two-dimensional fluid model for a number of frequencies in the range 4–40 MHz.

In the process of optimization CAPs sources, the competition between the creation of active neutral species (used for soft treatments) and ion species is critical for many applications when heating of the gas is undesired. Indeed, gas heating comes mostly by electron elastic collisions with the background gas and large electron densities are consequently unwanted. The design of CAPs will be then driven by the analysis of the production ratio between active neutral species Ar* and ionic species. One of the external plasma parameters which can be easily changed experimentally is the driving frequency. In this study, we will investigate the effect of f_d from a fundamental point of view while keeping in mind its experimental implications.

The model was described in details in [1] and has three structure units: *configuration*, *chemistry* and *transport*. For the sake of simplicity, the *configuration* we study here is the classical parallel plates case. The electric field between the electrodes is controlled externally by a voltage and responded internally by the space charge distribution. For the voltage we selected sinusoidal RF functions of different frequencies around $f_d^* = 13.56$ MHz. The amplitude is fixed at U = 300V. To compute the space charge distribution and the electric field we solve the Poisson's equation.

The species involved in the *chemistry* can be divided into background gas species and active species. The background gas is composed of Ar in ground state. The gas temperature is set to $T_g = 350$ K. Four active species are considered, namely argon excited atom (Ar^*), atomic (Ar^+) ion, molecular (Ar_2^+) ion and electron (e). The excited species are those of the 4s level block treated as a lumped level. The processes taken into account are elastic scattering (R0), excitation (R1), direct ionization (R2), stepwise (R3) and Penning (R4) ionization, molecular ion formation (R5), radiative decay (R6), dissociative recombination (R7) and diffusion (R8). To account for the deviations from Maxwell equilibrium we calculated the electron energy distribution function (EEDF) using the Boltzmann solver Bolsig+.

The *transport* part couples the transport of the relevant species (electrons, ions, excited atoms) to the electric field and electron energy balance. The governing fluid equations are the continuity equation for the excited atoms, ions and electrons, the transport equations of the considered particles solved in a drift–diffusion approximation and the energy equation for the electrons.

To investigate the results, we first deal with macroscopic effects like current and voltage from which we zoom into the underlying microscopic features; which are densities and reaction rates.

The effect of the driving frequency f_d on the plasma current I and voltage V is given in figure 1 for three f_d values namely $1/2\ f_d^* = 6.78$, $f_d^* = 13.56$ and $2f_d^* = 27.12$ MHz.

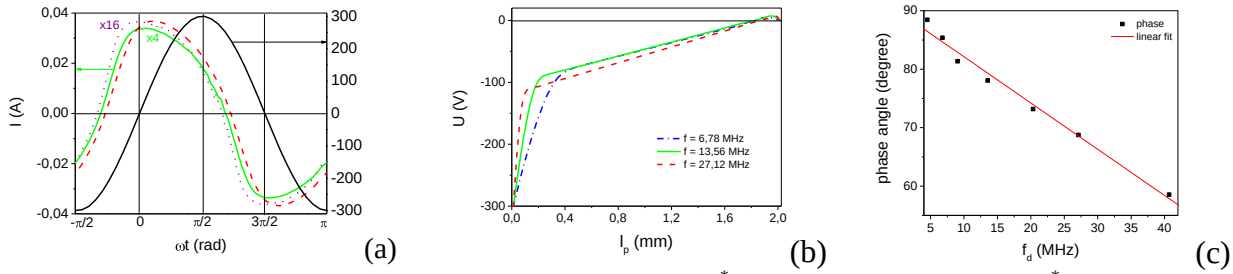

Fig. 1: (a) Plasma current in one cycle of the applied voltage for ½f_d^* = 6.78 (dotted purple line), f_d^* = 13.56 (straight green line) and $2f_d^*$= 27.12 MHz (dashed red line) together with the voltage phase distribution (black line); (b) voltage V(x) at phase π as a function of position x; (c) Voltage-current phase difference versus f_d.

The figure shows that, there is a quadratic dependence of the current amplitude I_{max} on the frequency. This dependance can be derived also analytically.

Much less dramatic is the change in the potential distribution V(x) as function of f_d as can be seen in figure 1(b) where V(x) is given for the same three f_d values; in all cases the left electrode is in the cathode phase of maximum negative potential whereas the right electrode is grounded. The figure shows that the change in the voltage drop ΔV_s over the sheath varies from 218 V at 6.78 MHz to 190 V at 27.12 MHz, which is about 13% or 6% per frequency step. The voltage drop over the plasma ΔV_p changes in a complementary way since the sum of the two voltage drops should equal the voltage offered by the power supply. From the current and voltage time dependence the phase angle is determined. It is observed that the overall change is modest; a change in f_d with a factor 10 only gives a variation of 30%. So the plasma remains capacitive but becomes (slightly) more resistive increasing f_d.

One of the main objectives of this study is the question how much the value of the driving frequency will change the ratio $n(Ar^*)$ / n_e. This dependence given in figure 2(a) shows that $n(Ar^*)$ / n_e goes down with increasing f_d. The heating aspect of the plasma increases with respect to the ability to generate radicals.

The increase of n_e due to the increasing f_d is reflected in the enhancement of the reaction rates. This can be observed in figure 2 where the reaction rates for f_d^* (fig. 2b) are compared with those of $2f_d^*$ (fig.2c). It is seen that due to the step of factor of 2 most of the reactions increase with more than one order. The highest change is that in the direct ionization (R2) which goes up with more than 2 orders. However, this is only relevant for a small region in the pre-sheath. The smallest change (factor 5) is that found for the radiation (R6), which is due to the fact that the excitation changes with less than one order while at the same time the radiation has to compete with stepwise ionization (R3). The latter, due to the f-doubling, has increased more or less 20 times. Although the excitation (R1), increases less than the other processes it stays three times higher than the rest. This implies that the Ar^* remains the dominant species. However, as it was shown the ratio $n(Ar^*)/n_e$ goes down increasing f_d. This is not always beneficial for practical applications, as more charges means more current conducted to the treated surface. Additionally higher n_e values will result in an increase of the gas heating whereas the (relative) decrease in $n(Ar^*)$ will lower the performance of the plasma in surface interactions.

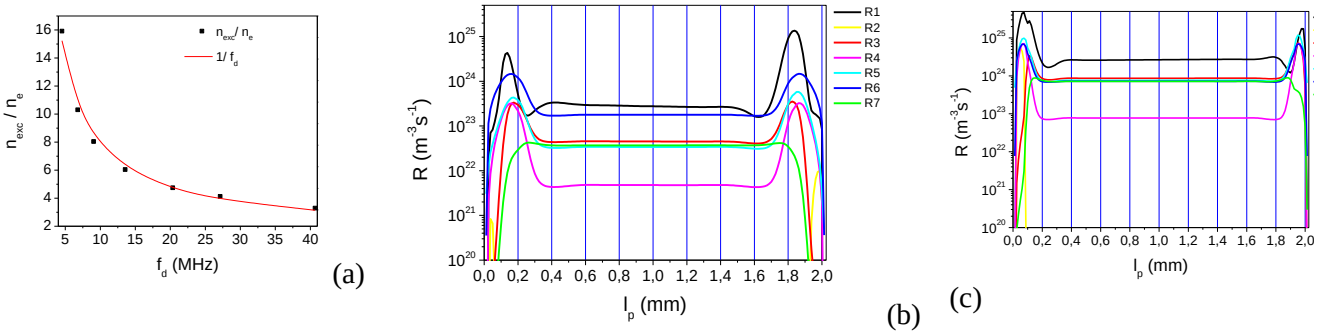

Fig. 2: Ratio of excited species over electron density as function of frequency (a); Spatial distributions of the reaction rates for f_d^* = 13.56 MHz (b) and $2f_d^*$ (c).

References

[1] M. Atanasova, E. A. D. Carbone, D. Mihailova, E. Benova, G. Degrez and J. J. A. M. van der Mullen *J. Phys. D Appl. Phys.* (2012) in press.